







Upgrading of composites.

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Abstract of EP0206832

Substrates such as chipboard or cement aggregate board are upgraded with a cured rubber layer by forming on the substrate a composite of uncured but curable rubber sheeting containing a heat activatable curative with an intermediate film of polar thermosetting resin, and hot pressing the uncured composite so as to form polar bonds between the resin and substrate with simultaneous co-vulcanisation of the rubber and the resin.

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㉑ **Upgrading of composites.**

㉑ Substrates such as chipboard or cement aggregate board are upgraded with a cured rubber layer by forming on the substrate a composite of uncured but curable rubber sheeting containing a heat activatable curative with an intermediate film of polar thermosetting resin, and hot pressing the uncured composite so as to form polar bonds between the resin and substrate with simultaneous co-vulcanisation of the rubber and the resin.

EP 0 206 832 A2

This invention relates to the upgrading of substrate elements by providing them with a layer of cured elastomeric material, and is particularly but not
5 exclusively concerned with composite elements which are useful in the building and construction industry by virtue of having a desired degree of weather resistance.

The principle of producing weather resistant panels or
10 mouldings having protective elastomeric layers is known from EP 9053 and CA 1 150 465, the teachings of which are incorporated herein by reference.

According to EP 9053, a process for producing weather resistant panels or mouldings which consist of wood
15 particles mixed with binding agents, of fibres containing lignocellulose or of raw materials prepared in another way and which are pressed together under the influence of heat, is characterised in that before hot pressing a covering layer made of vulcanisable elastomeric materials, such as natural and/or synthetic
20 rubber or the like, is applied to the topside and/or underside of the cake of chips which has been formed from the wood particles, fibres containing lignocellulose or raw materials prepared in another way, whereupon hot pressing is carried out with simultaneous
25 vulcanising of the covering layers. Panels or mouldings produced in this way are characterised by a permanently

1 elastic covering layer which is connected intimately to
the chip body and which is fully vulcanised. Such
panels or mouldings may be coloured and/or provided on
the outer surface of the elastic layer with grooves or
5 grains or other patterns. Furthermore, the elastic
layer may incorporate a reinforcement in the form of a
web or fabric.

According to the above prior art, many wood fibre
materials for the building industry are limited to
10 interior use since they are not sufficiently weather
resistant to be used externally. Such materials, for
example particle boards, fibre boards or insulating
boards are cheap, light and insulating and so are
ideal building components; however, a problem facing
15 the building industry has been the requirement of pro-
viding necessary weather resistance and ageing stability
to the materials. Modification by glues, hydrophobic
agents or antibacterial agents have not provided
realistic solutions from an economic or practical
20 viewpoint. More recent developments such as applying
to the materials layers of synthetic resin-soaked
papers, sheet metals, asbestos cement or synthetic
foils have also proved unsatisfactory, since these tend
to peel off the substrate under the influence of the
25 weather (temperature variations, radiation and moisture).

The solution proposed by the art specifically mentioned

1 above provides a building board which is said to be
economical to produce, weather resistant, substantially
non-ageing, embrittling or cracking, and which has high
abrasion-resistance, impact resistant capacities and
5 can be made substantially non-inflammable. Indeed, the
general teaching of the art is to provide in a single
production step a woodchip or lignocellulosic substrate
including a curable resin binding agent, coated with a
weather resistant elastomeric material, the elastomeric
10 material being in vulanised form and being both adhered
to one surface of the base material (substrate) and
penetrated and impregnated into that surface of the
base material. The art teaches the use of a natural
vulcanisable rubber or a synthetic vulcanisable rubber
15 as the elastomeric material. Such rubbers may also
include stabilising agents and further additives so as
substantially to reduce the brittleness and cracking
thereof under the influence of rain, ozone, light
radiation, UV radiation and heat radiation. The
20 elastomers may also be modified by altering the chemical
composition and the type and quantity of added fillers.
This, though, is as far as the prior art disclosure of
suitable elastomeric layers goes. It is said that the
essence of the prior art technique is the fact that the
25 elastomeric coating penetrates into the substrate wood
fibres in a single production run, without the need of
adhesive agents, thus the process comprises substantially
simultaneously compressing the particles of the woodchip

1 or lignocellulose substrate and hardening the adhesive
agents (binding agents) contained therein; and compres-
sing and vulcanising the covering layer of natural
rubber and/or synthetic rubber or other elastomer. The
5 elastomer coating may be applied by spraying, pouring,
sprinkling, painting, or it may be applied in the form
of a self-supporting film, that is a rubber sheet.

It will be appreciated therefore that the above-mentioned
patents describe the principle of coating woodchip in a
10 single simultaneous operation, but without giving full
details of the best means of obtaining such panels.

Further work on this topic has lead to certain selected
and advantageous developments of the basic art, but
again concerned with a simultaneous or one-step
15 production technique, whereby the elastomeric layer is
applied to a bed of chips or fibres and the whole
finished panel is produced in one operation. This
technique is useful, but it will be appreciated that
such a one-step method can only be used by producers
20 who have the facility for manufacturing the basic
substrate, that is chipboard or fibre board. A more
useful technique has been recognised as being the
upgrading of existing or already produced substrates
such as chipboard, concrete board or aggregate board.
25 The ability to provide a firmly adhering elastomeric
layer to such boards would provide a greater flexibility
to the production industry and also a wider variety of

1 products, since it is possible to apply elastomeric
layers to already formed substrates on a smaller scale
than if the substrate itself has to be produced in the
same operating step.

5 A detailed study of the so-called two-step or sequential
route to elastomer-coated boards has shown that
are many difficulties associated with the apparently
simple technique of adhering an elastomeric sheet to a
pre-formed substrate. Considerable expertise has been
10 found to be necessary in order to produce a reliable
product which remains integral on handling immediately
after its production, which may be efficiently produced on
a commercial scale and which remains in a usable
condition following the generally rough treatment it
15 may receive in use. Moreover, it is important that the
product should withstand the effects of weathering when
exposed to the atmosphere for many years. In particular,
the elastomeric outer layer of such products is prefer-
ably long-term UV stable and flame retardant
20 and should have good peel strength.

EP-A-0097279 teaches the hot pressing of a cover layer of
unvulcanised rubber onto a fibrous composition including a
synthetic resin and curative, with a thermoplastic resin
film intermediate these two layers. The product, intended
25 for car components eg. dust covers and in which the fibres
may be for example wool, cotton or glass wool, is not
proposed for building element use where long exposures to
high temperatures and hence structural integrity is a
requisite.

1 GB 1529778 teaches the production of laminated plastics
materials with non-slip surfaces by hot pressing a rubber
composition onto a core or substrate carrying a
heat-hardenable melamine-formaldehyde impregnated paper on
fabric, to form a consolidated product. The rubber
5 composition is a creamy mixture of chlorosulphonated
polyethylene rubber, the resin, and a curative, and because
of its nature is not self-supporting but has to be carried
on a temporary support during the production process, which
support is subsequently removed.

10

JP 53-69816 teaches the lamination of a decorative eg
rosewood layer onto a basic wood plate by first spreading
eg melamine resin onto the wood and applying an elastic
sheet thereto, followed by hot pressing, and thereafter
15 spreading the adhesive onto the sheet and applying the
decorative layer with further hot pressing. This is
clearly the simple use of melamine resin in its well-known
adhesive form, there being no teaching to employ same as a
self supporting film, or of its use with particulate
20 cellulosic substrate or with specific rubbers which
convulcanise with the melamine under hot press conditions.

According to the present invention, there is provided a
process for producing a vulcanised composite comprising a
25 substrate having a cured elastomeric layer secured thereto,

1 which process comprises (a) providing (1) a self-supporting
sheet of uncured elastomeric material comprising a
vulcanisable elastomer selected from copolymers of
ethylene, at least one other alpha olefin and from zero to
5 a minor proportion of at least one copolymerisable polyene,
butyl rubbers, chlorosulphonated polyethylene rubbers, and
mixtures of two or more thereof, and a heat activatable
curative system therefor, (2) a self-supporting film of
10 polar thermosetting resin and (3) a substrate comprising
cellulosic particles and being capable of chemical
interaction with said resin at elevated temperature; (b)
forming an uncured composite of the substrate and the sheet
with the resin film therebetween; and (c) hot pressing the
15 uncured composite to form polar bonds between the resin and
substrate and simultaneously to co-vulcanise the
elastomeric material and the resin to generate the desired
vulcanised composite.

20 The substrate employed in the process of the
invention comprises cellulosic particles and is preferably
wood chipboard or lignocellulosic fibre board. Such
materials are well-known in the building and furniture
industry, but principally for indoor use; without special
25 treatment, such boards do not have the necessary weather
resistance for outdoor use. Such boards are conventionally
produced by hot pressing of a bed of cellulosic particles
eg wood, fibres or chips, impregnated with a binding

- 1 agent. Preferably, such boards are hardened with a polar
resin binding agent, and it is preferred that this resin
should be a phenol-formaldehyde resin,
melamine-formaldehyde resin, isocyanate resin or
5 urea-formaldehyde resin. Substrates incorporating such
resins have been found to be particularly susceptible to
coating with elastomeric layers by the process of the
present invention.
- 10 The cellulosic particles of the substrate may alternatively
be bonded with inorganic material such as cement. It is
preferred that such cement compounds are in the form of
boards where the final product is intended for use in the
building industry. Vulcanised composites based on cement
15 bonded woodchip boards find use as external claddings in
the building industry. The provision of a cured
elastomeric layer for such boards means that they acquire a
weather resistance not hitherto possible when the materials
themselves are exposed to the atmosphere; such composites
20 may be generally thinner, lighter and of improved impact
strength, and hence cheaper and easier to handle than
conventional uncoated cement boards.

Of the elastomeric materials which may be employed in
25 accordance with the invention, it is particularly preferred
that the elastomeric material should comprise a copolymer
of ethylene and at least one other alpha-olefin and from 0
to a minor proportion of at least one copolymerisable
polyene. More preferably, the elastomeric material is a
30 terpolymer of ethylene, at least one other alpha-olefin

1 and a minor proportion of at least one copolymerisable
diene. Particularly preferred is an ethylene-propylene-
diene terpolymer (EPDM), for example containing from 1
to 10% of diene. The diene may be for example 1,4
5 hexadiene, dicyclopentadiene or an alkylidenenorbornene
such as methylenenorbornene or ethylidenenorbornene, or
vinylidenenorbornene, although the many alternatives
known in the art are suitable. Such elastomer, when
vulcanised, inherently has good weather resistant
10 properties, and by incorporating other components into
the elastomeric material, such properties can be
optimised for the envisaged end use of the vulcanised
composite. Also preferred as elastomer is the EPM copolymer rubber.
It is particularly preferred that the sheet of uncured vulcanisable
15 elastomer should be a mixture of EPDM and EPM copolymer rubber, the
proportions being adjusted on a cost basis as necessary.

The curative system contained in the elastomer sheet is
selected to provide curing action on the elastomer,
and will be one which is activated at the temperature
20 at which hot pressing is conducted. Preferably the
curative system is a peroxide or peroxide-based system,
and this is particularly advantageous in the case where
the elastomeric material is EPDM or EPDM/EPM blend,
since EPDM cured via peroxide gives a particularly
25 hard-wearing and weather resistant vulcanised product.
More preferably, the peroxide system comprises aromatic
or aliphatic peroxides such as the aromatic diacyl
peroxides and aliphatic diacyl peroxides, dibasic acid

1 peroxides, ketone peroxides, alkyl peroxyesters, alkyl
hydroperoxides, eg diacetylperoxide, dibenzoyl-peroxide,
bis-2, 4-dichlorobenzoyl peroxide, ditert.-butyl
peroxide, dicumylperoxide, tert.-butylperbenzoate,
5 tert.-butylcumyl peroxide, 2,5-bis-(tert.-butylperoxy)-
2,5-dimethylhexane, 2,5-bis-(tert.-butylperoxy)-2,5-
dimethylhexyne-3; 4,4,4', 4(-tetra-(tert.-butylperoxy-
isopropyl)-benzene, 1,1-bis-(tert.-butylperoxy)-3,3,5-
trimethylcyclohexane, lauroyl peroxide, succinic acid
10 peroxide, cyclohexanone peroxide, ter.-butyl peracetate
and butyl hydroperoxide. The peroxide is preferably
present in the elastomeric material in an amount of
from 2-20, more preferably from 5-15 parts by weight of
peroxide per 100 parts by weight of vulcanisable
15 elastomer(phr). Such proportions of curative have been
found to give excellent covulcanisation of the elastomeric
material and the film of polar thermosetting resin.
It is this co-vulcanisation, coupled with the bonding
of the resin to the substrate, which is believed
20 to give the good adhesion properties possible by use of
the process of the invention. As will be appreciated
by those skilled in the art, other curative systems are
possible, depending on the desired end properties and
the nature of the elastomer, for example, sulphur or a
25 phenol resin curative system may be used.

- 1 The elastomeric material may alternatively or additionally
comprise a butyl rubber (including halogenated butyl
rubbers) or a chloro-sulphonated polyethylene rubber. Such
materials, in the cured state, have certain properties
5 which make them suitable for outdoor applications, but are
in general less preferred than EPM and/or EPDM rubbers.

It is a particular advantage of the defined process that
the elastomer, before the hot pressing stage, is in the
10 form of a self-supporting sheet. This greatly facilitates
commercial scale production since the elastomer can be
delivered to the substrate surface, after application of
the resin film, from a drum or roll. In contrast, rubber
formulations in cream form which have to be carried on a
15 temporary support are difficult to handle and do not
readily lend themselves to large, commercial scale
production techniques.

The self supporting film of polar resin is a heat curable,
20 ie. thermosetting, resin, such as methacrylate,
polyurethane or epoxy resin. However, it is especially
preferred that the resin comprises melamine eg.
melamine-formaldehyde or a phenolic resin.

- 25 The resin is applied to the substrate in the form of a
self-supporting film, and this brings with it the
production conveniences mentioned above with regard to the
self-supporting elastomer sheet.

1 In the case of melamine resin, it is particularly preferred
for the film to comprise so-called melamine paper, that is
paper or tissue impregnated with melamine resin.

Preferably the film comprises from 20 to 200, more
5 preferably about 200 parts by weight of resin per 100 parts
by weight of paper or tissue, since this heavier weight of
melamine paper has been found to give exceptionally good
bonding between the elastomer sheet and the substrate.

10 The conditions under which hot pressing is performed
will depend upon the nature of the elastomer and resin
systems employed. However, typically, hot pressing
will be carried out at a temperature of from 130 to
250°C, more preferably from 160 to 220°C. Similarly,
the pressure applied to the composite will be dependent
15 on the systems employed, but typically from 1000 to
10000 kPa, more preferably from 2000 to 6000 kPa. For
example, hot pressing may be carried out for a period
of 0.5 to 15 minutes, which is generally sufficient to
convert the uncured composite to a fully vulcanised
20 material in which the elastomer layer is firmly adhered
to the substrate.

Investigations have shown that the process of the
invention may be performed on hot press equipment which
25 is conventionally employed to upgrade chipboard with

1 rigid plastic sheeting, even though the elastomer sheet
itself is not rigid. Such equipment typically
comprises a bed on which the substrate board is laid,
and a hot press face which descends on the substrate
5 after the required over-layer has been applied thereto.
In such equipment, after hot pressing, the pressure is
generally released, and the board is removed from the
bed immediately, that is whilst the composite is still
hot, by means of the application of vacuum cups to the
10 upper surface of the composite.

The process of the present invention can be carried out on
such equipment, and it is to facilitate good and efficient
commercial operation that the resin film is required to be
15 thermosetting. If the resin were to be thermoplastic, then
the vulcanised composite would have to be allowed to cool
below the plastic limit of the resin, before the pressure
could be released. If the pressure were to be released
whilst the system is still hot, then the vulcanised
20 elastomer sheet would pull away or bulge from the
under-lying substrate due to the presence of moisture and
air voids in the substrate, whether this be chipboard,
lignocellulosic fibre board or cement chipboard. The
applied

1 temperature, for example 180°C, generates steam or
other vapour in the system if pressure is released, and
this causes physical deformation of the sheet if thermo-
plastic resins are used.

5 Similarly, in use of conventional
equipment wherein the hot pressed composite is lifted
away from the hot press bed by means of vacuum cups, if
the resin film which is intermediate the elastomer and

10 substrate is still plastic, the suction effect pulls the
sheet away from the substrate. The resin used in
accordance with the process of the invention is therefore
required to be thermosetting, and it will be appreciated
that with such materials, once the thermoset temperature
15 has been reached, the resin becomes solid and hence is not
susceptible to bulging or lifting away from the substrate
by virtue of pressure being released prior to cooling, or
by virtue of the application of suction to the upper
20 surface of the applied elastomeric sheet. In other words,
thermosetting resins, especially melamine, does not give
hot flow in the composite.

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1 It has been found that melamine, particularly in the form
of melamine paper, performs particularly well in
conjunction with EPDM or EPDM/EPM blend as elastomer
and peroxide as the curative system. Without wishing
5 to be bound by theory, it is believed that, under hot
press conditions, melamine cures by a condensation
reaction, and the free radicals generated in the EPDM
via the peroxide curative give good chemical bonding
with the condensing melamine molecules. Simultaneously,
10 the application of pressure to the system forces the
melamine into voids and interstices in the substrate,
especially chipboard, so giving a physical linkage
between the EPDM or EPDM/EPM sheet and the substrate.
Moreover, it is believed that the polar nature of the
15 resin leads to chemical inter-action with the resin
present in the chipboard (as binding agent in the
original production process therefor). In the case of
cement bonded chipboards, this will already contain
molecules, eg silica, which are polar in nature.

20

The hot pressing operation is preferably continued
until the elastomeric material is fully cured, since
this gives optimum weather resistance to the finished
product. Moreover, in order to obtain good adhesion
25 between the elastomer and the substrate, the hot
pressing is also preferably performed so that the

1 intermediate thermosetting resin film is also fully cured.
This, in conjunction with the preferred peroxide
curative system will optimise the strength of adhesion to
the substrate. Although
5 for certain end uses a peel strength in excess of,
say, 0.3 kg/cm may be acceptable, the hot pressing is
preferably carried out until the resulting composite
has a peel strength of the sheet on the substrate of at
least 1 kg/cm, as measured in accordance with ASTM
10 D816. It is particularly preferred that for building
industry end uses, the elastomer/ curative/polar resin
film/ substrate system be selected in conjunction with
applied hot pressing conditions so that a final peel
strength in excess of 2 kg/cm is obtained in the
15 finished vulcanised composite.

As mentioned previously, the particular characteristics
of EPDM make this especially suitable for composites
designed for outdoor applications. The inherent
properties of EPDM or any particular elastomer employed
20 in accordance with the invention may be modified by the
inclusion of other additives such as those selected
from UV stabilisers, flame retardants, plasticizers,
reinforcers, fillers, heat stabilisers, coupling
agents, processing aids, colouring agents and acidity
25 buffers in any combination.

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1 It will be appreciated that selection of these additives
depends on the proposed end use of the upgraded substrate.
For example when the rubber is black (contains carbon
black) then no extra UV-stabilizer package will be
5 necessary, but if the rubber contains whitener eg
TiO₂ or other colouring agents, then it may be
desirable to incorporate UV stabilizers or acidity
buffers. Again, depending on envisaged end use, the
rubber may contain a flame retardant filler package or
10 reinforcement fillers.

The substrate to which the elastomeric material is
applied in accordance with the invention may be of
substantially planar format, or it may be pre-moulded
into a shaped configuration, to suit the required end
15 use. In addition, it is possible to apply the elasto-
meric material thereto in accordance with the invention
such that the elastomer forms a type of hinge to allow
movement of adjacent parts of the substrate. The
elastomer sheet may be applied simply to one surface of
20 the substrate, although in other embodiments the resin
film and elastomer sheet may be applied, in step (b) of
the process, to more than one surface of the substrate
prior to hot pressing. Thus composites in which both
their inner and outer surfaces have an elastomeric
25 facade are possible. In order to make the composites
more attractive, the elastomeric material may include

1 additives which provide colour thereto. In addition,
or in the alternative, the elastomeric sheet may have,
or may be provided during the hot pressing step with, a
grooved, grained or otherwise patterned outer surface,
5 to suit the required end use of the composite.

In a particularly preferred aspect, the process according
to the invention provides for the upgrading of woodchip
board or cement bonded woodchip board by providing the same
with a firmly adhering elastomeric layer, the process
10 comprising forming an uncured composite of the board, a
self supporting sheet of uncured EPDM or EPDM/EPM blend
rubber containing a peroxide curative system, and,
intermediate said board and sheet, a self-supporting film
of a thermosetting resin which is co-vulcanisable with said
15 EPDM or EPDM/EPM at elevated temperature, and hot pressing
said uncured composite to form polar bonds between the
resin and the board and simultaneously to co-vulcanise the
EPDM and the resin. All the relevant preferred aspects of
the process as described hereinbefore, apply to this
20 specific aspect of the invention. In the case where
the composite to be upgraded is chipboard, it is
preferred to use melamine or phenolic resin as the
polar resin intermediate; in the case of cement bonded
chipboard, it has been found that use of methacrylate,
25 polyurethane or epoxy resins brings certain advantages.

Examples

- 1 A vulcanisable elastomeric composition having the
formulation (in parts by weight) as listed in Table 1 was
prepared by the following technique. First the filler and
plasticizer components were mixed in an internal mixer (15
5 litre capacity, Banbury type) at 40-50°C. After 30 seconds'
mixing, both elastomeric polymers (EPM and EPDM) were added,
together with all other components except the curatives.
The mixing chamber was then closed and the mixing was
continued with heating of the chamber until after 4-5
10 minutes the temperature had reached $150^{\circ}\text{C} \pm 10^{\circ}\text{C}$. At this
stage the mixture was dumped on to a mill and sheeted out to
a thickness of 1.5 cm, the sheets then being allowed to cool
to room temperature.
- 15 In a second pass through the internal mixer, the sheet was
introduced into the chamber at 40-50°C and the curatives
were added. Mixing and heating was continued over 1-2
minutes until a temperature of 90-100°C had been reached.
The mixture was then dumped onto the mill and sheeted to a
20 thickness of 1.5 cm, followed by cooling to room
temperature. Thereafter the sheet was calendered to a
desired thickness of 1.0 mm, the rubber at this stage still
not being in the cured state, but being intended for curing
in a subsequent hot pressing stage when applied to a
25 chipboard. However, to determine the physical properties of
the final product coating, a sample of the thus-formed
sheeting was press cured at 50kg/cm^2 , 180° for 8 minutes and
the cured product was tested

- 1 for physical and cure properties, as representative of the nature of the cured rubber when adhered to a substrate in accordance with the invention.
- 5 The cure characteristics of the sheet compound, as measured on a Monsanto oscillating rheometer of arc ± 5 at 180°C, were as follows:

	ML minimum	21	pound/inches
10	MH maximum	116	pound/inches
	ts ₂ scorch	0.35	minutes
	tc ₉₀ cure	1.05	minutes

The characteristics of the press cured sheeting were as follows:-

	Shore A hardness	80	ASTM D 2240
	100% modulus (MPa)	4.4	ASTM D. 412
	300% modulus (MPa)	-	"
20	Tensile strength (MPa)	5.6	"
	Elongation at break (%)	233	"

TABLE 1

<u>Composition</u>	<u>1</u>
(1) EPDM	25
(2) EPM	75
(3) Filler (a)	180
(4) Filler (b).	20
(5) Plasticizer	20
(6) Stabilizer (a)	40
(7) Stabilizer (b)	0.3
(8) Stabilizer (c)	0.15
(9) Process aid (a)	10
(10) Process aid (b)	1
(11) Process aid (c)	3
(12) Activator	1
(13) Zinc oxide	5
(14) Coupling agent	2
(15) Curative (a)	14

1 The components used in producing the above composition were
as follows:

- (1) EPDM = VISTALON 7000 of Essochem, an
5 ethylene/propylene/diene terpolymer
rubber of Mooney ML(1+8) at 127° C =
55, 70 wt% ethylene content and high
diene content
- 10 (2) EPM = VISTALON 504 of Essochem, an
ethylene/propylene copolymer rubber of
Mooney ML (1+8) at 127° C = 25; 50 wt%
ethylene content
- 15 (3) Filler (a)= Martinal OL 111 Aluminium trihydroxide
- (4) Filler (b)= KS 300 reinforcing silica filler
- (5) Stabilizer= diiso decyl phthalate (DIDP)
- 20 (6)-(8) Stabilizers (a),(b),(c) = titanium dioxide,
Tinuvin 770 and Irganox B215 UV
stabilizers, respectively

- 1 (9)-(11) Process aids (a),(b),(c) = Allied Chemicals low
density polyethylene ACPE 617A; stearic
acid; and polyethylene glycol 4000
respectively
- 5
- (12) Activator = trialkylcyanurate, a peroxide activator
- (13) Zinc Oxide
- 10 (14) Coupling Agent = Union Carbide vinyl silane Al72, for
coupling fillers and polymers
- (15) Curative (a) = Trigonox 29/40 MB of Akzo (1,1-di-tert
butyl-peroxy - 3,3,5 trimethyl cyclohexane
15 in a masterbatch with EPDM, peroxide content
40wt%.

- 1 The uncured elastomeric sheeting produced as described
above was used to upgrade wood chipboard by a hot press
technique, using self-supporting resin films as the means
for securing adhesion and covulcanisation in the finished
5 product. The substrate employed was a preformed
commercially available woodchip board having a polar resin
bonding system and being 18 mm in thickness. Samples of
the elastomeric sheeting were applied to the substrate with
a variety of self-supporting resin films as intermediate
10 layer, and the uncured composite was then subjected to hot
pressing under time, temperature and pressure conditions
which permitted covulcanisation of the elastomer and the
intermediate resin, with formation of polar bonds between
the resin and substrate. In fact for each sample the hot
15 pressing was conducted for 2 - 3 minutes at 200° C and 35
kg/cm². The pressure was released at the elevated
temperature, and the samples were allowed to cool to room
temperature.
- 20 The samples of vulcanised composite produced by the above
technique were cut to strips 6.5 cm wide and subjected to a
standard peel test in accordance with ASTM D816, the
measurements being made at room temperature and at 180° C
on an Instron tensile tester. The results obtained are
25 shown in Table 2, where the values for samples produced in
accordance with the invention are listed numerically and
comparisons are alphabetical.

Table 2

<u>Sample</u>	<u>Resin Film</u>	<u>Thickness</u> Micron	<u>Peel Strength</u> (kg/cm)	
			20°C	180°C
1	Melamine	50	3.2	1.2
2	Phenol	50	3.8	1.4
A	EAA	50	4.4	0.5
B	EAA	50	3.7	0.5
C	EVA	50	3.2	0.5
D	Ionomer N22	50	3.4	0.5
E	Ionomer N21	50	3.1	0.5
F	LDPE	50	3.7	0.5
G	LLDPE	50	4.6	0.6

The films, each 50 microns thick, used in the preparation of these samples were as follows:

- 1 = Melamine HP paper, an impregnated melamine paper of total weight (per sheet) 128 g and containing 92 g thermosetting melamine resin.
- 2 = Phenolic Kraft paper, being (per sheet) 80 g paper impregnated with 120 g thermosetting phenol resin.
- A = Ethylene/acrylic acid copolymer (EAA) TR 5100 of Essochem, containing 12 wt% AA and having melt index (ASTM D 1328) = 8.0 g/10 min and melting point 98.5°C

- B = EAA TR 5000 of Essochem containing 5 wt% AA and having melt index (ASTM D 1328) = 8.0 g/10 min and melting point 103° C
- C = Ethylene/vinyl acetate copolymer (EVA) Escorene Ultra UL 00018 of Essochem containing 18 wt% VA and having melt index (ASTM D 1328) = 0.05 g/10 min.
- D = Ionomer produced by crosslinking EAA of sample A with a sodium salt.
- E = Ionomer produced by crosslinking EAA of sample A with a zinc salt.
- F = Low density polyethylene Escorene LD 180 of Essochem having melt index (ASTM D 1328) = 2.0 g/10 min.
- G = Linear low density polyethylene Escorene LL1001 of Essochem having melt index (ASTM D 1328) = 1.0 g/10 min.

It will be appreciated that the resin films of samples A - G are thermoplastic and therefore the process used to produce such samples does not fall within the scope of the invention. The resin films (papers) of samples 1 and 2 are however thermosetting, so the process was within the scope of the invention.

1 Referring to Table, 2 it is seen that the room
temperature (20° c) peel strength values for all the
samples were approximately the same, in the 3.5-4.5
range, and such values are perfectly acceptable for
5 construction elements which are only exposed to room
type temperatures.

However, the values measured at 180° C clearly show
the great advantage of using thermosetting resin
papers in the process compared with the conventional
10 thermoplastic films. The samples produced by the
process of the invention had peel strengths some 2 to
3 times those of the comparisons, and this is of
importance in construction elements which are exposed
15 to the environment: temperatures considerably above
100° C can readily be reached in many parts of the
world, and even in relatively temperate climates, and
it is of importance that building facades or other
structural elements which might comprise the
20 vulcanised composites produced by the process of the
invention do not disintegrate in these conditions.

CLAIMS

1. A process for producing a vulcanised composite comprising a substrate having a cured elastomeric layer secured thereto, which process comprises (a) providing (1) a self-supporting sheet of uncured elastomeric material comprising a vulcanisable elastomer selected from copolymers of ethylene, at least one other alpha olefin and from zero to a minor proportion of at least one copolymerisable polyene, butyl rubbers, chlorosulphonated polyethylene rubbers, and mixtures of two or more thereof, and a heat activatable curative system therefor, (2) a self-supporting film of polar thermosetting resin and (3) a substrate comprising cellulosic particles and being capable of chemical interaction with said resin at elevated temperature; (b) forming an uncured composite of the substrate and the sheet with the resin film therebetween; and (c) hot pressing the uncured composite to form polar bonds between the resin and substrate, and simultaneously to co-vulcanise the elastomeric material and the resin to generate the desired vulcanised composite.
2. A process according to claim 1, wherein the substrate comprises wood chipboard, lignocellulosic fibre board or cement bonded wood chipboard.

3. A process according to claim 2, wherein the wood chipboard or fibre board has been hardened with a polar resin binding agent comprising phenol-formaldehyde, melamine-formaldehyde, isocyanate or urea-formaldehyde resin.
4. A process according to claim 1, 2 or 3 wherein the elastomeric material comprises EPM copolymer rubber, EPDM terpolymer rubber or a blend of EPDM and EPM.
5. A process according to any one of the preceding claims, wherein the curative system is peroxide or peroxide-based.
6. A process according to claim 5 wherein the peroxide or peroxide-based curative system is present in an amount of from 5 to 15 parts by weight per 100 parts by weight of the elastomeric material.
7. A process according to any one of the preceding claims wherein the thermosetting resin film comprises a methacrylate resin, a phenolic resin, an epoxy resin or a polyurethane resin.
8. A process according to any one of claims 1 - 6 wherein the thermosetting resin film comprises melamine.

9. A process according to claim 8 wherein the thermosetting resin film comprises paper or tissue impregnated with melamine resin.
10. A process according to any one of the preceding claims, wherein hot pressing is carried out at a temperature of from 130 to 250° C.
11. A process according to any one of the preceding claims, wherein hot pressing is carried out at a pressure of from 1000 to 10000 kPa.
12. A process according to any one of the preceding claims, wherein the uncured composite is subjected to pressure for a period of from 0.5 to 15 minutes.
13. A process according to any one of the preceding claims wherein, on completion of hot pressing, the pressure is released whilst the vulcanised composite is still hot.
14. A process according to claim 13, which comprises the additional step of lifting the vulcanised composite by means of a vacuum cup applied to the composite after release of pressure but whilst said vulcanised composite is still at elevated temperature.

15. A process according to any one of the preceding claims, wherein the cured elastomeric layer is substantially fully vulcanised.
16. A process according to any one of the preceding claims, wherein hot pressing is carried out until the resulting vulcanised composite has a peel strength of the sheet on the substrate of at least 1 kg/cm (ASTM D 816).
17. A process according to claim 16 wherein hot pressing is carried out until the resulting vulcanised composite has a peel strength of the sheet on the substrate of at least 2 kg/cm (ASTM D 816).
18. A process according to any one of the preceding claims, wherein the elastomeric material includes additives selected from UV stabilisers, flame retardants, plasticizers, reinforcers, fillers, heat stabilisers, coupling agents, processing aids, acidity buffers, colouring agents and mixtures of two or more thereof.
19. A process according to any one of the preceding claims, wherein step (b) is conducted by applying the resin film and elastomer sheet to more than one surface of the substrate prior to hot pressing.

20. A process according to any one of the preceding claims, wherein the elastomeric sheet has, or is provided during the hot pressing with, a grooved, grained or otherwise patterned outer surface.
21. A building, a construction, a constructional element or an architectural feature which comprises or incorporates a vulcanised composite produced by the process according to any one of the preceding claims.